

10/519521  
DT01 Rec'd PCT/PTC 23 DEC 2004

**AMENDMENTS TO THE SPECIFICATION**

Please replace the paragraph at page 1, beginning on line 5, with the following rewritten paragraph:

--The present invention relates to a method for manufacturing a high silicon grain-oriented electrical steel sheet that can improve magnetic properties, especially, a core loss characteristic, and more specifically, to a method for manufacturing a high silicon grain-oriented electrical steel sheet in which a powder coating agent containing an annealing separator for siliconization is coated on the surface of a steel sheet, and finished annealed to thereby ~~manufacture~~provide an electrical steel sheet with outstanding high frequency magnetic properties as well as outstanding commercial frequency properties.--

Please replace the paragraph at page 2, beginning on line 2, with the following rewritten paragraph:

--Recently, as electrical devices are diversified, demands on devices operating in a high frequency band ~~increase~~have increased and thus ~~desires on demand for a~~ core material with superior magnetic properties in high frequency has also ~~start to~~ ~~increase~~increased.--

Please replace the paragraph at page 3, beginning on line 15 and ending on page 4, line 2, with the following rewritten paragraph:

--~~As prior arts known as~~Among the prior art methods ~~that can for the~~ manufacture of high silicon steel sheets, Japanese Patent Laid Open Publication No. S56-3625, ~~etc.,~~ discloses a direct casting of a high silicon steel using a single roll or twin rolls, Japanese Patent Laid Open Publication No. S62-103321, ~~etc.,~~ discloses a warm rolling in which rolling is performed in a ~~heating~~heated state ~~of at~~ a proper temperature, and Japanese Patent Laid Open Publication No. H5-171281, ~~etc.,~~ discloses a clad rolling in which rolling is performed in a state ~~that~~wherein the high silicon steel is located at an inner portion and a low silicon steel is located at an outer portion. However, the aforementioned prior ~~arts have~~ ~~been~~art has not yet been commercialized.--

Please replace the paragraph at page 4, beginning on line 2, with the following rewritten paragraph:

--For mass production of high silicon steel products such as 3% Si non-oriented steel products, ~~there is~~ a well known ~~a process including~~ includes the steps of depositing silicon on a surface of a material by a chemical vapor deposition using  $\text{SiCl}_4$  and then homogenizing the silicon, as disclosed in ~~prior arts of~~ Japanese Patent Laid Open Publication No. S62-227078, US Patent No. 3,423,253 and the like. However, the above process causes the ~~produced~~ manufactured products to be sold inevitably at a price five times high than the conventional 3% Si steel products due to the difficulty in the CVD process. In spite of the fact that ~~the produced~~ these products have superior magnetic properties, it is difficult to popularize and commercialize such products due to the high cost.--

Please replace the subheading on page 5, line 7 with the following rewritten centered subheading.

~~--DISCLOSURE SUMMARY OF THE INVENTION--~~

Please replace the heading on page 6, line 19 with the following rewritten centered heading.

~~--BEST MODE FOR CARRYING OUT~~ DETAILED DESCRIPTION OF THE  
INVENTION--

Please delete the sentence on page 6, line 19.

Please replace the paragraph at page 6, beginning on line 20 and ending on page 7, line 19, with the following rewritten paragraph:

--The manufacturing processes of the grain-oriented electrical steel sheet have differ somewhat ~~differences~~ according to manufacturers. However, each of the processes generally includes the steps of: adjusting the contents of components in the steel making process; producing a casting slab; reheating the casting slab; hot rolling the reheated casting slab; annealing the hot rolled sheet and cold rolling the hot-rolled steel sheet so as to adjust the thickness of the steel sheet; decarburization annealing the cold-rolled steel sheet; high

temperature annealing the steel sheet for a secondary recrystallization; and finish coating an insulating film on the steel sheet. The above process is based on mass production. In the mass production, it is an important factor to establish a production facility toward the cold rolling. Then, as aforementioned, higher silicon content in the electrical steel sheet decreases core loss, magnetostriction, coercive force, and magnetic anisotropy but increases maximum permeability, thereby demonstrating excellent magnetic properties. However, since the elongation that is a kind of mechanical properties abruptly decreases depending on an increase in silicon content, it is known that up to 3.3% Si is contained in a starting material to which the cold rolling enabling the mass production of electrical steel is applicable.--

Please replace the paragraph at page 7, beginning on line 20 and ending on page 8, line 12, with the following rewritten paragraph:

--Accordingly, the inventor has researched processes for manufacturing high silicon electrical steel sheets by using a conventional electrical steel sheet manufacturing process employing the cold rolling, which enables mass production. As a result, the inventor has found that a grain-oriented electrical steel sheet with ~~very~~-excellent magnetic properties can be manufactured by a process comprising the steps of: preparing slurry formed by dispersing a powder coating agent in water or the like, the powder coating agent being made by mixing ~~an~~ a sintered powder of Fe-Si group having a predetermined grain size and Si content with MgO powder as the annealing separator; coating the prepared slurry on a surface of a decarburized and nitrogen-annealed electrical steel sheet; diffusion annealing the resultant steel sheet during the high temperature annealing process to complete a high silicon content and magnetic properties by a second recrystallization, and suggests the present invention.--

Please replace the paragraph at page 8, beginning on line 13 and ending on page 9, line 4, with the following rewritten paragraph:

--That is, in the present invention, in order to prevent sticking between materials while a high temperature annealing for secondary recrystallization is performed to manufacture a conventional grain-oriented electrical steel sheet, an annealing separator is inevitably coated on a surface of a steel sheet. At this time, the annealing separator is coated

in a state that an Fe-Si-based sintered powder group having a predetermined grain size and Si content is added to MgO powder as main component of the annealing separator, so that a high silicon grain-oriented electrical steel sheet can be manufactured through a subsequent high temperature annealing process. In other words, the present invention ~~can produce~~provides a high silicon grain-oriented electrical steel sheet with ~~very~~-excellent magnetic properties while employing the conventional process for manufacturing grain-oriented electrical steel sheet using the cold rolling.--

Please replace the paragraph at page 9, beginning on line 5, with the following rewritten paragraph:

--First, the inventive siliconizing powder coating agent will be described in ~~concrete~~detail.--

Please replace the paragraph at page 9, beginning on line 7, with the following rewritten paragraph:

--When contacting silicon (Si) with Fe metal under high temperature hydrogen or nitrogen atmosphere more than 950 °C, there occurs an interdiffusion reaction where Si atoms diffuse into the Fe metal and Fe atoms diffuse into the Si containing metal ~~to make~~making the concentration of Fe and Si in both sides ~~to be~~-identical. Accordingly, when contacting Si metal powder on a matrix portion of the electrical steel sheet and then annealing the electrical steel sheet at a high temperature, an interdiffusion reaction may be progressed by an intermovement of metal Si and matrix Fe because the concentration of the Si powder is considerably higher than the Si concentration of 3% level in the surface of the grain-oriented electrical steel sheet.--

Please replace the paragraph at page 9, beginning on line 20 and ending on page 10, line 9, with the following rewritten paragraph:

--When comparing the interdiffusion reaction of Fe atoms and Si atoms, since the diffusion rate of Si atoms is faster by approximately two times than that of Fe atoms in a temperature range of 1000 - 1200 °C, ~~there occurs~~ a phenomenon called the Kirkendall effect occurs, corresponding to a non-homogeneous diffusion state. This non-homogeneous

diffusion state causes non-homogeneous state defects at a reaction interface or creates various compounds such as  $\text{FeSi}_2$ ,  $\text{FeSi}$ ,  $\text{Fe}_5\text{Si}_3$  and  $\text{Fe}_3\text{Si}$ , which act as a factor deteriorating magnetic properties. Accordingly, in a case where only the metal Si powder is used as the siliconizing agent, it is in fact impossible to produce high silicon grain-oriented electrical steel sheets having a homogenous composition without surface defects through a high temperature diffusion annealing.--

Please replace the paragraph at page 10, beginning on line 10, with the following rewritten paragraph:

--To solve the above problem, the inventor repeated researches on diffusion principle and so forth using Si powder and Fe powder, and finally found that the defects in the diffusion reaction portion are based on faster diffusion rate of Si than Fe. ~~Accordingly, the inventor suggests the present invention.--~~

Please replace the paragraph at page 10, beginning on line 16 and ending on page 11, line 4, with the following rewritten paragraph:

~~--That is, the~~The present invention is characterized by controlling grain size and composition of Si-containing powder agent used as the siliconizing agent so as to suppress the Si diffusion relative to the Fe diffusion. In other words, the present invention is characterized by providing an Fe-Si-based sintered powder controlled to have a predetermined grain size and composition to enable diffusion where Si atoms and Fe atoms are substituted with each other by an identical amount nearly without forming a composite compound where Fe and Si are bonded to each other at a diffusion reaction portion of the steel sheet surface, mixing the provided powder with annealing separator of  $\text{MgO}$  powder to form a mixture, and utilizing the mixture as the siliconizing coating agent.--

Please replace the paragraph at page 11, beginning on line 5, with the following rewritten paragraph:

--Hereinafter, the above characteristics will be described in ~~more~~concretegreater detail.--

Please replace the paragraph at page 11, beginning on line 7, with the following rewritten paragraph:

--First, to further ~~slack~~lower the diffusion rate of Si component, powder containing only Si metal is not used but Fe-Si-based compound such as  $\text{FeSi}_2$ ,  $\text{FeSi}$ ,  $\text{Fe}_5\text{Si}_3$  and  $\text{Fe}_3\text{Si}$ , ~~that wherein~~ Si metal is bonded to Fe metal is used as the main composition of the siliconizing coating agent.--

Please replace the paragraph at page 11, beginning on line 12 and ending on page 12, line 12, with the following rewritten paragraph:

--Fe-Si-based powder used in the present invention can be manufactured by mixing Fe powder and Si powder with each other, and sintering the mixed powder at a temperature range of 1000 - 1200°C in a mixture gas atmosphere of hydrogen and nitrogen for 5 - 10 hours, but is not necessarily restricted thereto and can be manufactured by various methods. At this time, the component ratio of the sintered powder compound is changed depending on the mixed amount of Fe powder and Si powder. Theoretically, when the mixed amount is 50%Si+50%Fe, the compound of  $\text{FeSi}_2$  is created, when the mixed amount is 34%Si+66%Fe, the compound of  $\text{FeSi}$  is created, when the mixed amount is 25%Si+75%Fe, the compound of  $\text{Fe}_5\text{Si}_3$  is created, and when the mixed amount is 14%Si+86%Fe, the compound of  $\text{Fe}_3\text{Si}$  is created. However, in actual annealing, small amounts of several compounds may exist according to an initial mixing state. In particular, when an annealing reaction is generated by a mixing of Fe powder and Si powder, the reaction ~~is progressed~~progresses in such a manner that Si atoms and Fe atoms are interdiffused to invade. Hence, although the amount of Si is somewhat large, the annealed powder ~~becomes~~ reaches a state in which surfaces of the sintered powder contain most of the  $\text{FeSi}_2$  compound or  $\text{FeSi}$  compound corresponding to a state that Fe atoms have been diffused exist but pure Si atoms exist at inside of the sintered powder. Accordingly, most of Fe-Si-based compound ~~exist~~ is present in the surface of the sintered powder.--

Please replace the paragraph at page 12, beginning on line 13 and ending on page 13, line 7, with the following rewritten paragraph:

--In the present invention, the Si content in the Fe-Si-based sintered powder obtained as above is restricted to 25 - 70 wt%. If the Si content is less than 25wt%, it is so small and thus diffusion rate may be very slow. Also, the high density of the annealed powder may cause ~~the~~ a drop of the dispersion when the coating process is performed in practice. Since the content of Si exceeding 70wt% allows the main component to exist as  $\text{FeSi}_2$  and a mixture of extra metal Si phase, the metal Si component contacts with the surface of material to increase the ~~creation~~ possibility of creating defects on the surface during the siliconizing process so that the control of the silicon content as siliconized may be difficult. In other words, by restricting the Si content contained in Fe-Si-based sintered powder to a range of 25 - 70 wt%, it is possible to manufacture Fe-Si-based composite compound sintered powder having  $\text{FeSi}_2$ , FeSi,  $\text{Fe}_5\text{Si}_3$  or  $\text{Fe}_3\text{Si}$  as a main component. It is more preferable that the content of  $\text{FeSi}_2$ +FeSi among the Fe-Si-based composite compounds should be restricted to 90wt% or more with respect to the total weight of the sintered powder.--

Please replace the paragraph at page 13, beginning on line 8, with the following rewritten paragraph:

--When Fe-Si-based sintered powder manufactured as above is mixed with MgO powder and is used as the coating agent of electrical steel sheet, ~~this.~~ This mixed powder is made in a slurry state and coated on the surface of the steel sheet by using a roll coater, which is most economical in commerical production ~~state~~. The Fe-Si-based sintered powder as the siliconizing agent should be made as fine as possible, ~~which.~~ This enhances the coating workability in a ~~production stage~~ commercial operation and is advantageous in terms of management of surface shape on the diffusion reaction. However, since the Fe-Si-based sintered powder where annealing reaction is completed is in a state of fused lump by a high temperature and long term reaction, it is necessary to control the grain size of the powder as fine as possible.--

Please replace the paragraph at page 13, beginning on line 22 and ending on page 14, line 7, with the following rewritten paragraph:

--Accordingly, the present invention ~~makes~~ requires that the grain size of the Fe-Si-based sintered powder ~~finely~~ be fine considering such a circumstance. ~~Finer~~ A finer

grain enhances the dispersity toward slurry state and improves the coatability. Also, by coating fine Fe-Si-based sintered powder on the surface of steel sheet, the surface contact area between the matrix material and the metal powder, i.e., interreaction area can be reduced to 30% or less compared with a single sheet contact. It is desirable that the grain size should be restricted to -325 mesh upon considering the productivity and costs for formation of fine powder.--

Please replace the paragraph at page 14, beginning on line 8, with the following rewritten paragraph:

--Also, the ~~inventive-final~~ powder coating agent is prepared by mixing Fe-Si-based sintered powder obtained as above with MgO powder ~~of which serves as an~~ annealing separator. Specifically, the inventive powder coating agent is prepared by mixing 100 ~~part~~ parts by weight of MgO, which is the main component of the annealing separator, with 0.5 - 120 ~~part-parts~~ parts by weight of the Fe-Si-based sintered powder. At this time, if the added amount of the sintered powder is less than 0.5 parts, the silicon content as siliconized is few or too small. If the added amount exceeds 120 parts, the dispersity of the sintered powder with MgO is poor, so that it is difficult to control the dispersity with MgO powder and to control the silicon content as siliconized according to the region of the matrix material, which is undesirable.--

Please replace the paragraph at page 16, beginning on line 2, with the following rewritten paragraph:

--Considering securing the hot rolling property and the magnetic properties, the steel slab is reheated at a temperature range of 1150 - 1340 °C, and is then hot rolled so that a hot rolled steel sheet with a thickness of 2.0 - 2.3 mm is made. Afterwards, hot rolled annealing is performed at a temperature below 1100 °C, and ~~picking~~ pickling and cold rolling are performed to control the thickness of the steel sheet to a range of 0.20 - 0.30 mm that corresponds to a final thickness. In case of 0.2 mm products, twice hot rolled annealing and cold rolling are performed to control the thickness of the steel sheet to the final thickness. After that, under a moisture atmosphere containing hydrogen and nitrogen, a decarburizing and nitriding treatment is performed at an approximate temperature range of 840 - 890 °C to



obtain a ~~decarburization~~ decarburized and nitrided annealed steel sheet. The aforementioned steps are well known in the conventional art, and the invention is not limited only to these ~~concrete~~ above described process conditions.--

Please replace the paragraph at page 16, beginning on line 20 and ending on page 17, line 3, with the following rewritten paragraph:

--The invention utilizes the decarburized steel sheet as the matrix steel sheet, which has a thin oxide layer formed on a surface thereof. Then, the thin oxide layer acts as a hindrance or barrier layer ~~of to the~~ interdiffusion reaction during the siliconizing annealing process and functions to decrease the amount of Si atoms diffused toward the inside of the matrix steel sheet. Accordingly, this thin oxide layer may be more advantageous in manufacturing an electrical steel sheet with superior core loss characteristics.--

Please replace the paragraph at page 17, beginning on line 4, with the following rewritten paragraph:

--Specifically, Fe-Si-based composite compound sintered powder is mixed with MgO powder to prepare the powder coating agent. The powder coating agent is dispersed in water and is made in a slurry-state. After that, the slurry coating agent is coated on the surface of the decarburized and ~~nitridation~~ nitrided annealed steel sheet by using a roll coater. At this time, the coating amount of the slurry coating agent is determined by the following formulas 1 and 2:--

Please replace the paragraph at page 19, beginning on line 25, and ending on page 20, line 1, with the following rewritten paragraph:

--Hereinafter, the present invention will be described in more detail with certain presently preferred embodiments. It will be understood that the below described embodiments are in no way limiting on the scope of the present invention.--

Please delete the heading on page 27, line 20.

Please replace the paragraph at page 27, beginning on line 21 and ending on page 28, line 4, with the following rewritten paragraph:

--As described above, although utilizing the conventional general manufacturing process, the present invention can coat a siliconizing coating composition on steel sheet instead of the conventional MgO composition as an annealing separator prior to the finish high temperature annealing and siliconize the coated siliconizing coating composition to manufacture a grain-oriented electrical steel sheet having superior magnetic properties and a thickness of 0.2 - 0.30 mm at a low production cost.--